Facile Synthesis and Catalysis of Pure-Silica and Heteroatom LTA

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Supporting Information

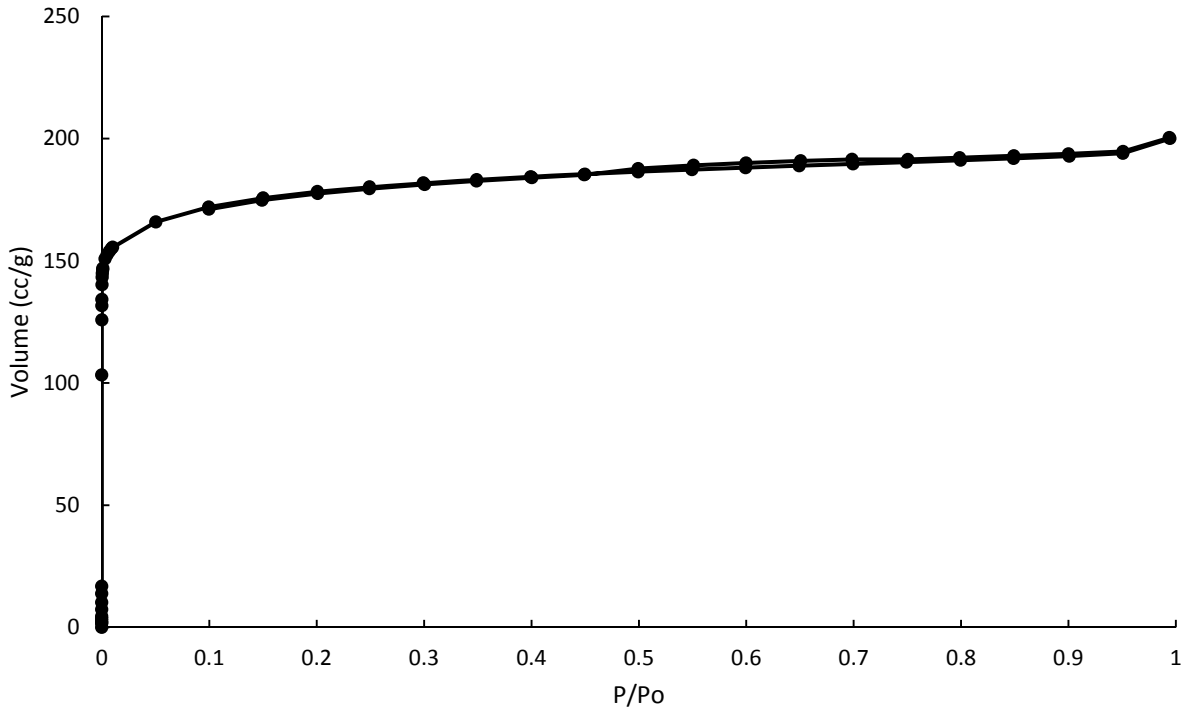

Figure S1. Nitrogen adsorption isotherm of calcined pure-silica LTA (t-plot micropore volume of 0.25 cc/g)

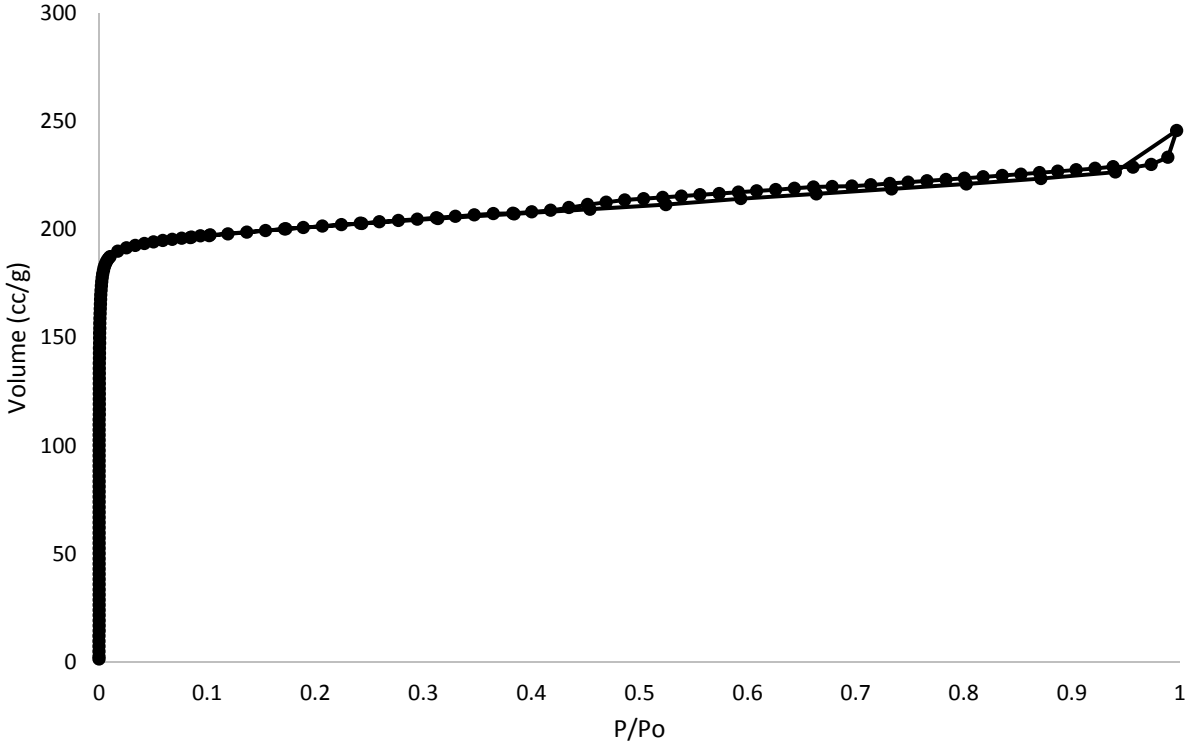

Figure S2. Argon adsorption isotherm of calcined pure-silica LTA

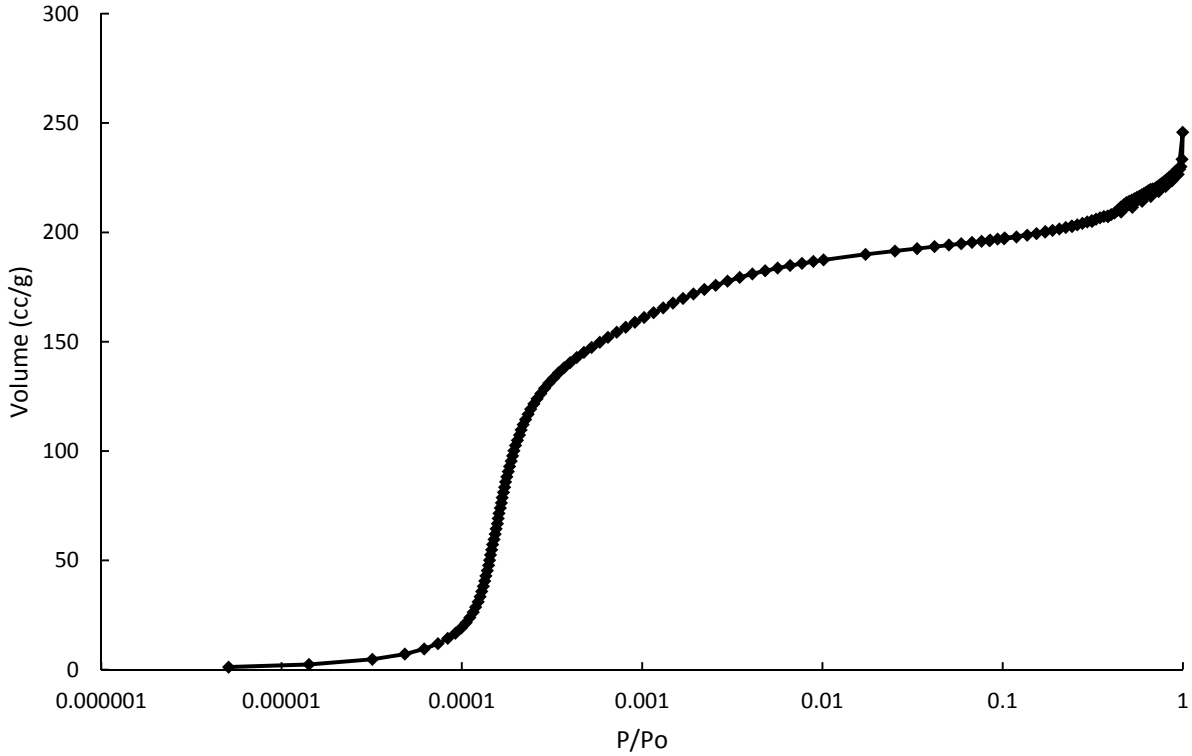

Figure S3. Log plot argon adsorption isotherm of calcined pure-silica LTA

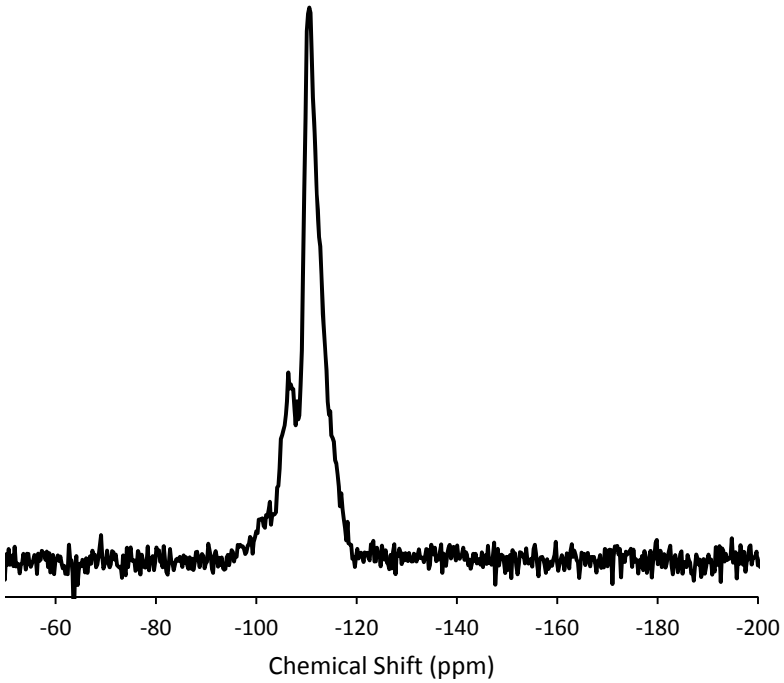

Figure S4. ^{29}Si CPMAS NMR of as-made Si-LTA

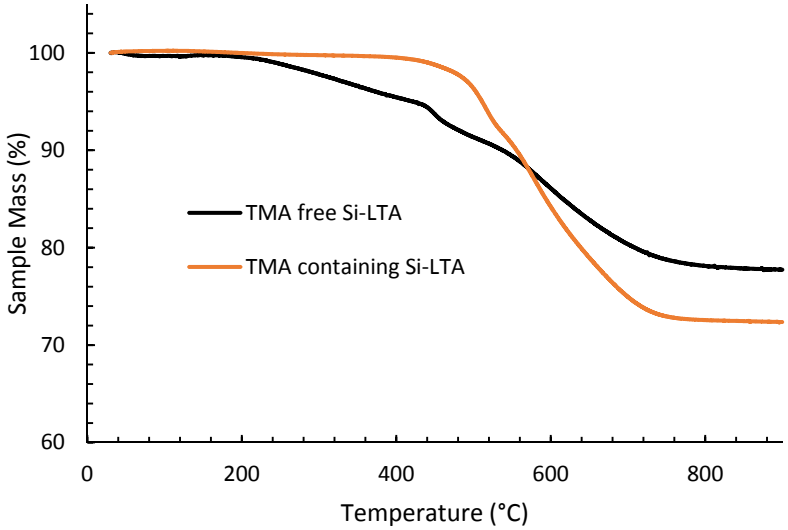

Figure S5. TGA analysis of as-made pure-silica LTA made with and without TMA

Molecular Modelling

We calculated the stablization energy of the OSDA reported here as well as that of juloidine in LTA. We used the IZA all-silica structure of LTA in these calculations. The stabilization energy is the difference in energy of the zeolite with occluded OSDAs and the isolated zeolite and OSDAs. It is reported on a kJ/(mol Si) basis. The stabilization energy is calculated by first placing *n* OSDAs per unit cell such that there is minimal steric overlap between the OSDA and the zeolite framework atoms. The system is then energy minimized. Then, 30 ps of molecular dynamics at 443K are carried out. The last 5 ps of the trajectory are used to compute an average energy of the system. This same calculation is carried out for the isolated zeolite and the isolated OSDA. All computations were conducted using the previously reported method.[1] The Dreiding forcefield was used in the GULP molecular modeling program to calculate the stabilization energies.

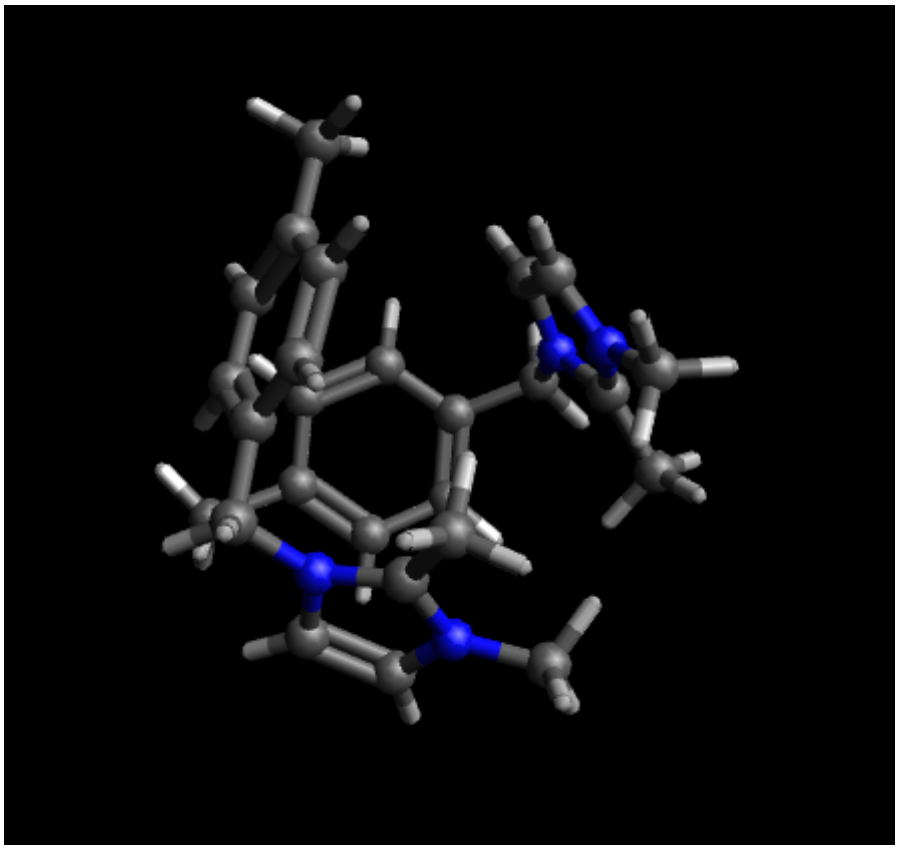

Figure S6. Conformation of the organic inside the α-cage of LTA for an occupany of 2 per cage. If only a single molecule of monoquats is occluded the stabilization energy value is -7.36 kJ/(mol Si).

The crystal structure of methylated julolidine was previously solved from single crystal analysis of the pure OSDA.[2] In the structure solution the molecules were found to dimerize with methyl groups pointing towards each other, so it has always been assumed that this was the conformation in the pore. Using molecular modelling we performed the molecular dynamics calculation of the stabilization energy for occluded julolidine 128 separate times. The results of those simulations are shown in Table S2 and images of the three different conformations are shown in Figure S7, Figure S8 and Figure S9. The molecular modelling results show that the most energetically favorable conformation is when the methyl groups point away from each other, not towards as was found in the single crystal. This result highlights the influence of the framework on the conformation of the organic. If only a single molecule of julolidine is occluded the energy is -6.35 kJ/(mol Si).

Table S1. Molecular modelling results for julolidine occluded in ITQ-29 (two per cage)

		Methyl Group Conformation		
Stabilization Energy (kJ/mol Si)	Total	Away	Towards	Opposite
average	-13.03	-13.80	-12.61	-12.85
upper	-14.27	-14.27	-14.05	-13.19
lower	-11.46	-12.66	-11.46	-12.21
median	-12.96	-14.05	-12.65	-12.87
# occurrences		33	30	69

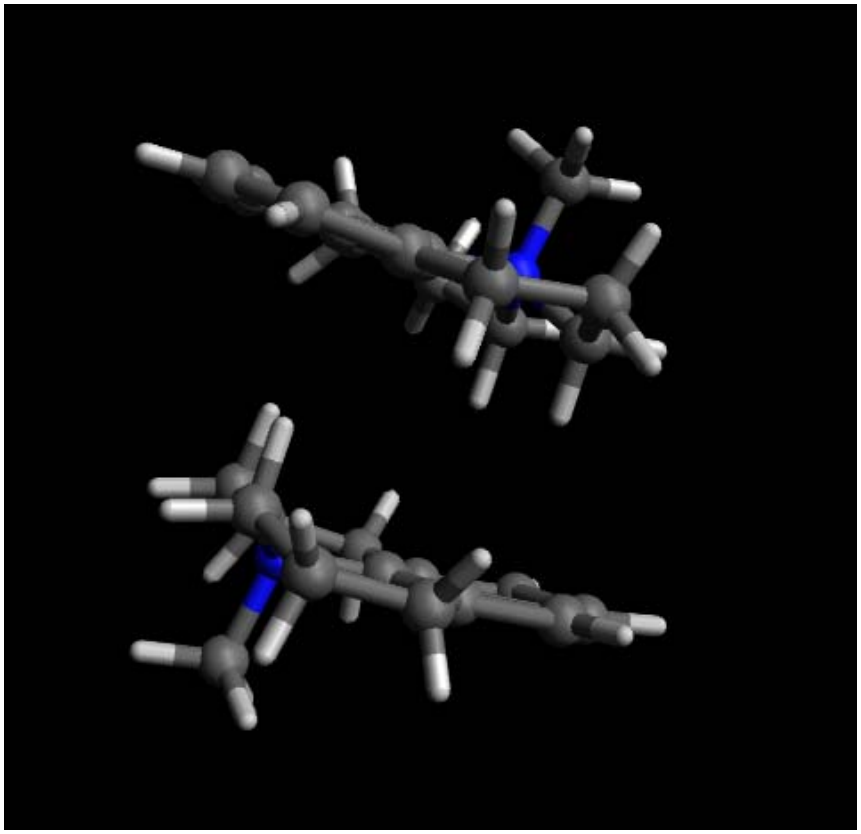

Figure S7. “Away” conformation of the organic in the α-cage of LTA

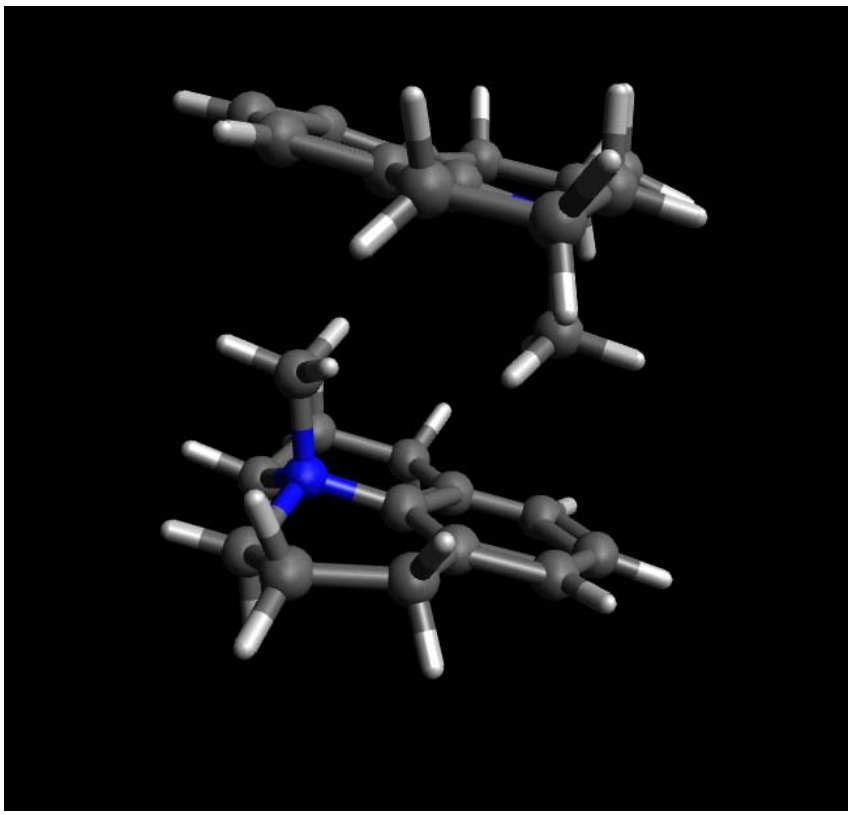

Figure S8. “Towards” conformation of the organic in the α-cage of LTA

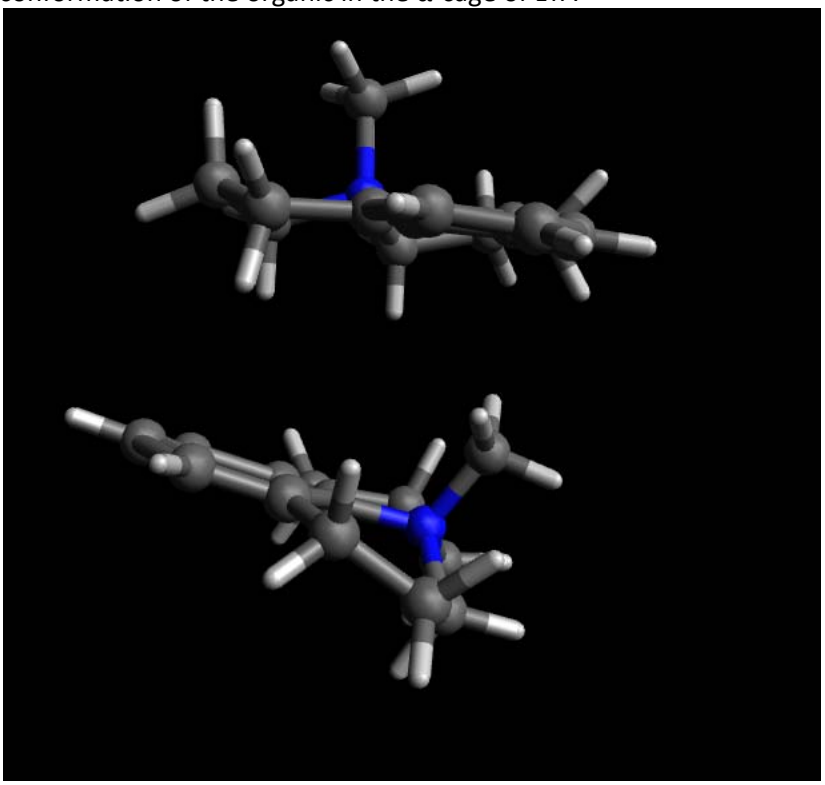

Figure S9. “Opposite” conformation of the organic in the α-cage of LTA
New OSDA average energy found to be -16.9 kJ/mol

Aluminum NMR

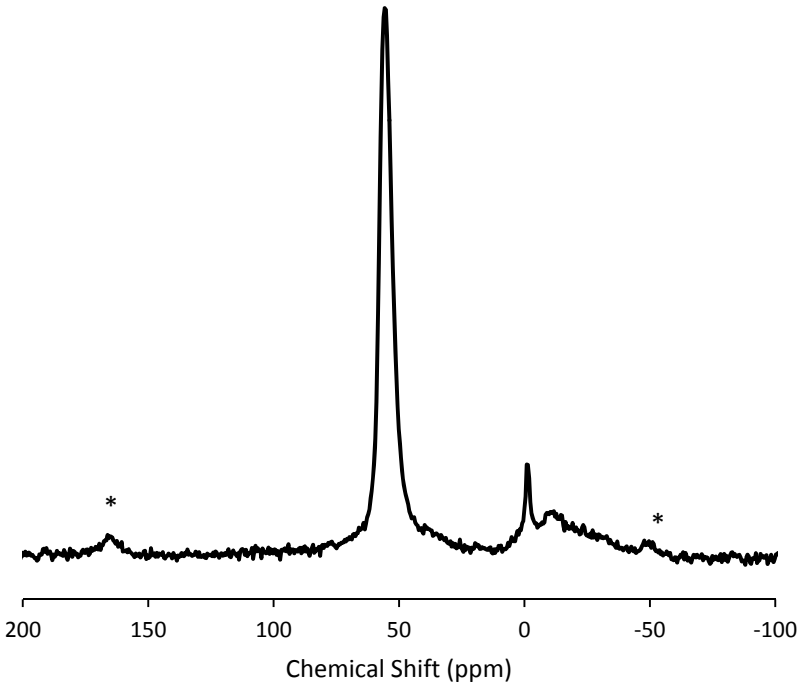

Figure S10. ^{27}Al NMR of calcined aluminosilicate LTA with gel Si/Al=20. Spinning sidebands are marked with *.

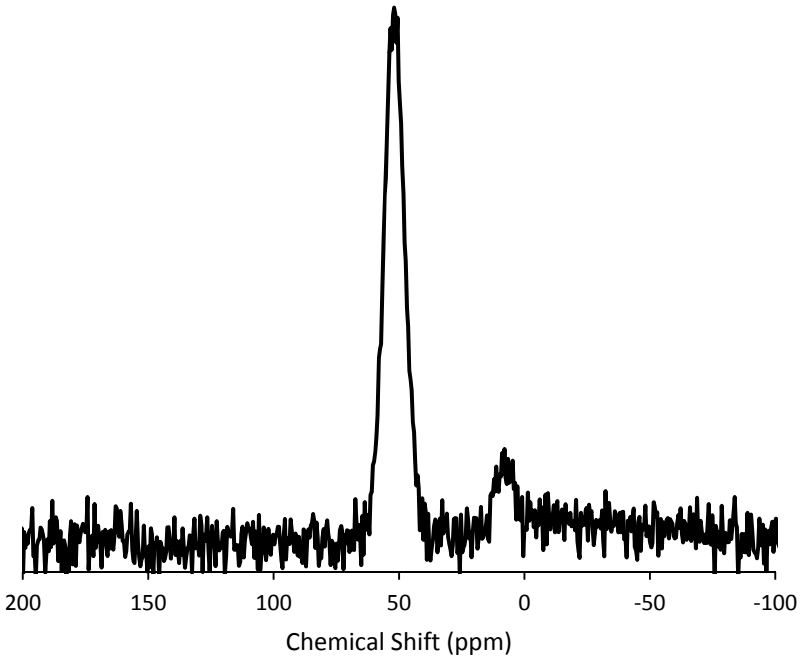

Figure S11. ^{27}Al NMR of as-made aluminosilicate LTA with gel Si/Al=20.

MTO reaction testing

Aluminosilicate LTA samples were also tested for their performance in the methanol-to-olefins (MTO) reaction. Prior to reaction testing, samples were calcined in breathing-grade air by initially drying them at 150 °C for 3 h (at a heating rate of 1 °C/min) before heating the samples further to 580 °C for 6 h (again at a 1 °C/min heating rate) to convert them to their proton forms. Calcined samples were then pelletized, crushed, and sieved to obtain particles between 0.18 mm and 0.60 mm, with approximately 200 mg of these pure zeolite particles being loaded into a 3/8" tubular, stainless steel reactor for each reaction test.

Reaction testing was conducted at 400 °C and atmospheric pressure, with a feed of 10% methanol in an inert gas blend (5% Ar, 95% He) for a total weight hourly space velocity (WHSV) of 1.3 h^{-1}. The reactor effluent gas was analyzed by an online GC/MS (Agilent GC 6980/MSD5793N), and conversion and selectivity data for each sample were computed on a carbon mole basis.

MTO Results:

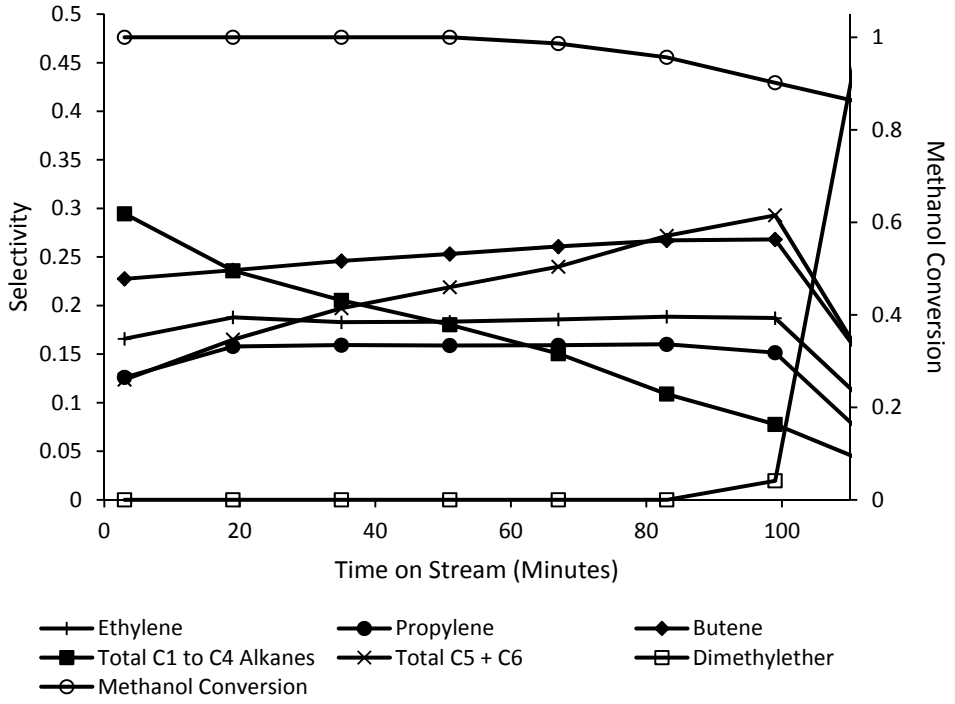

Figure S12. LTA with Si/Al=12

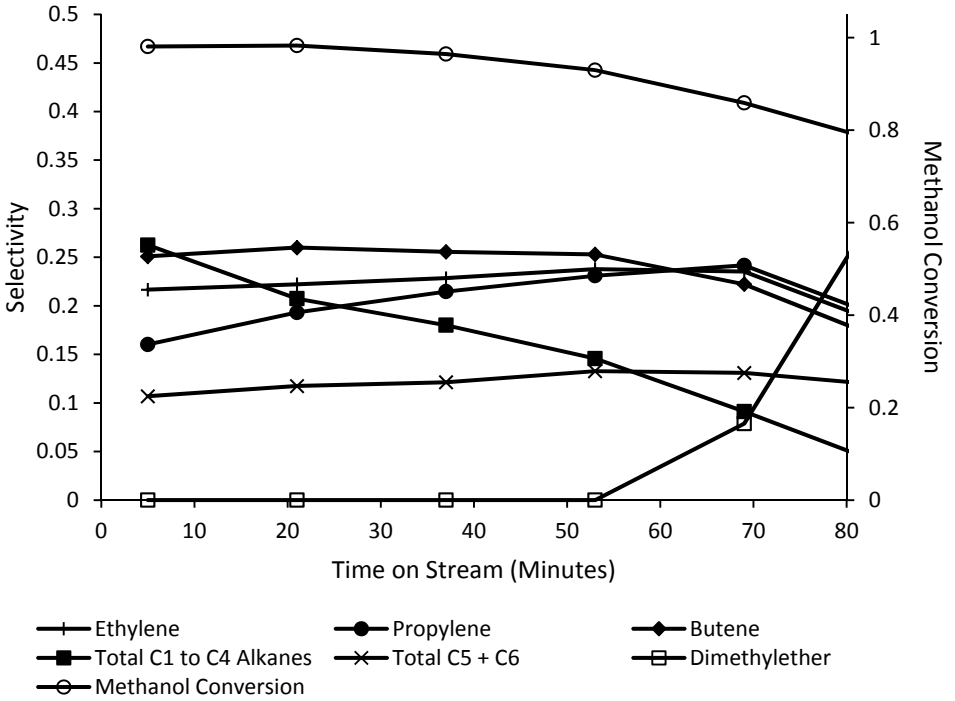

Figure S13. LTA with Si/Al=33

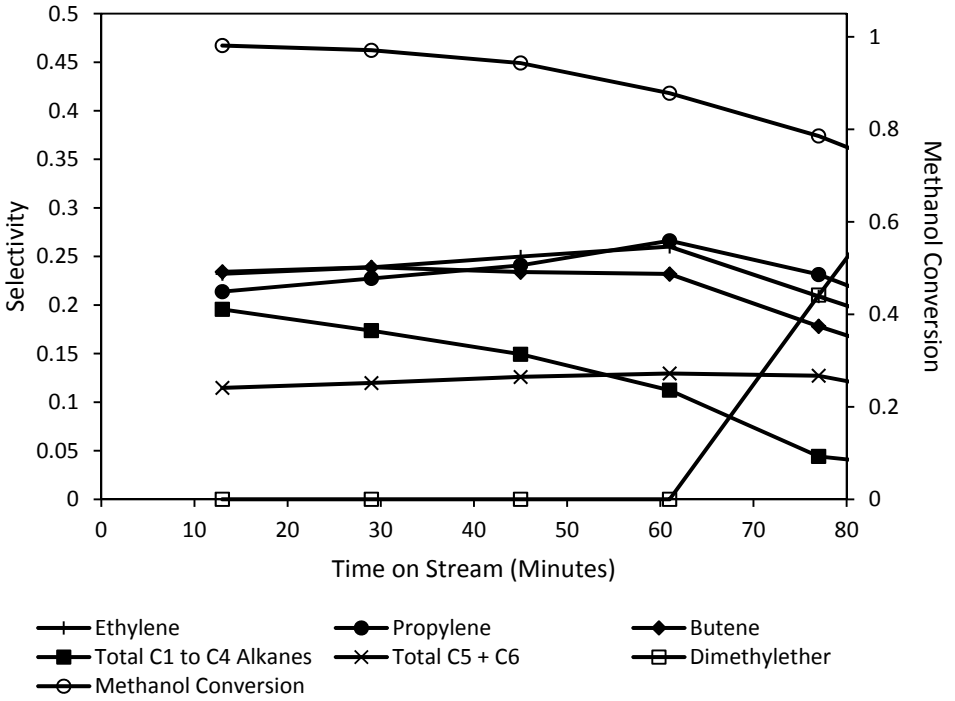

Figure S14. LTA with Si/Al=38

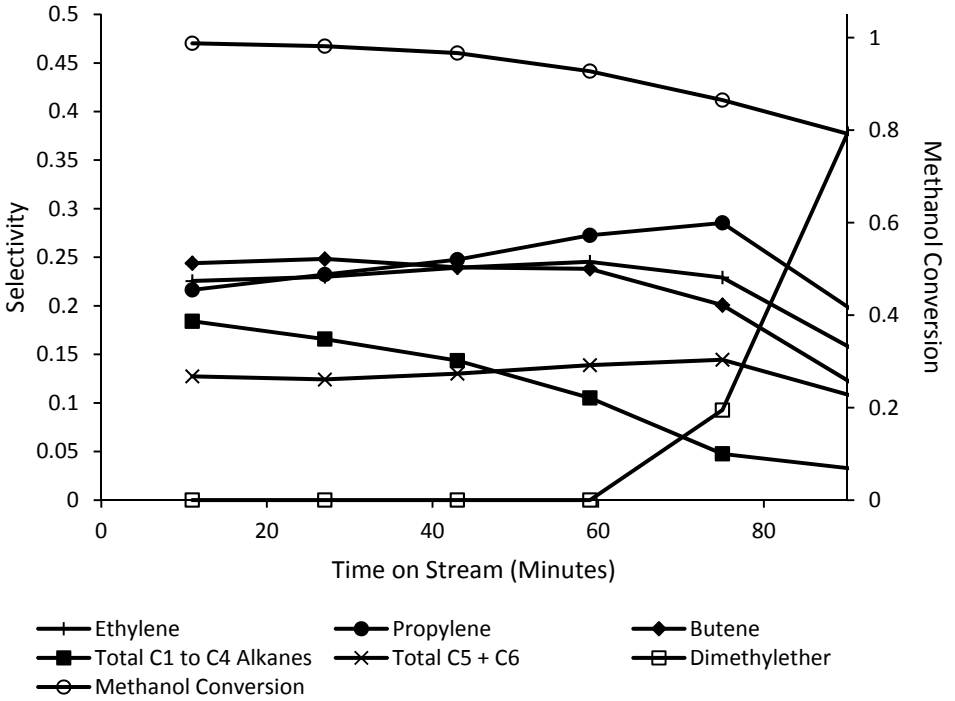

Figure S15. LTA with Si/Al=42

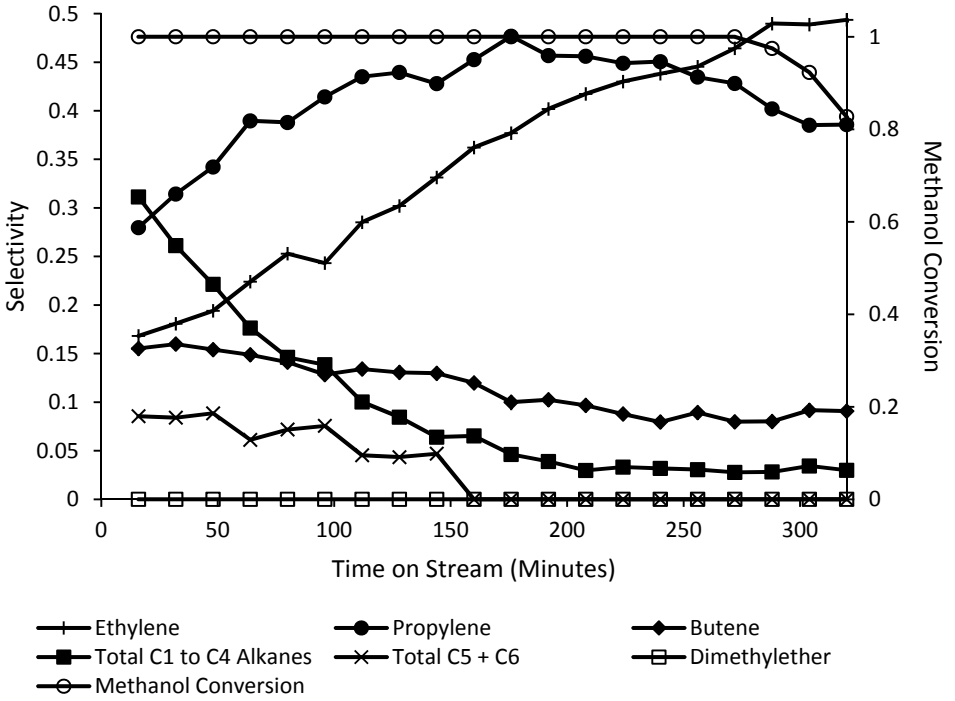

Figure S16. SSZ-13 (Si/Al=19)

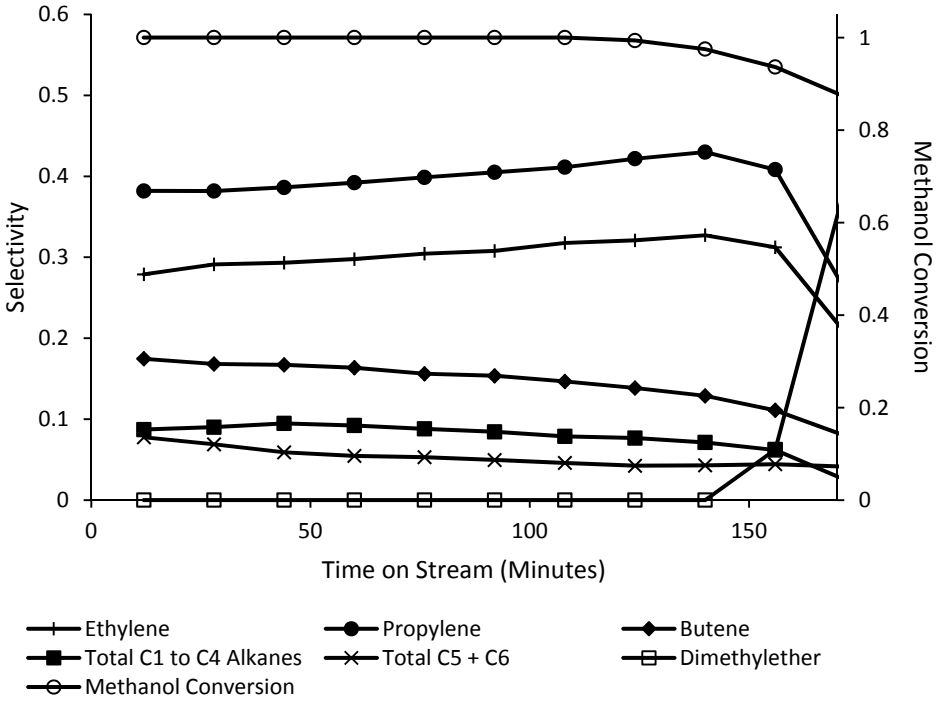

Figure S17. SAPO-34 MTO

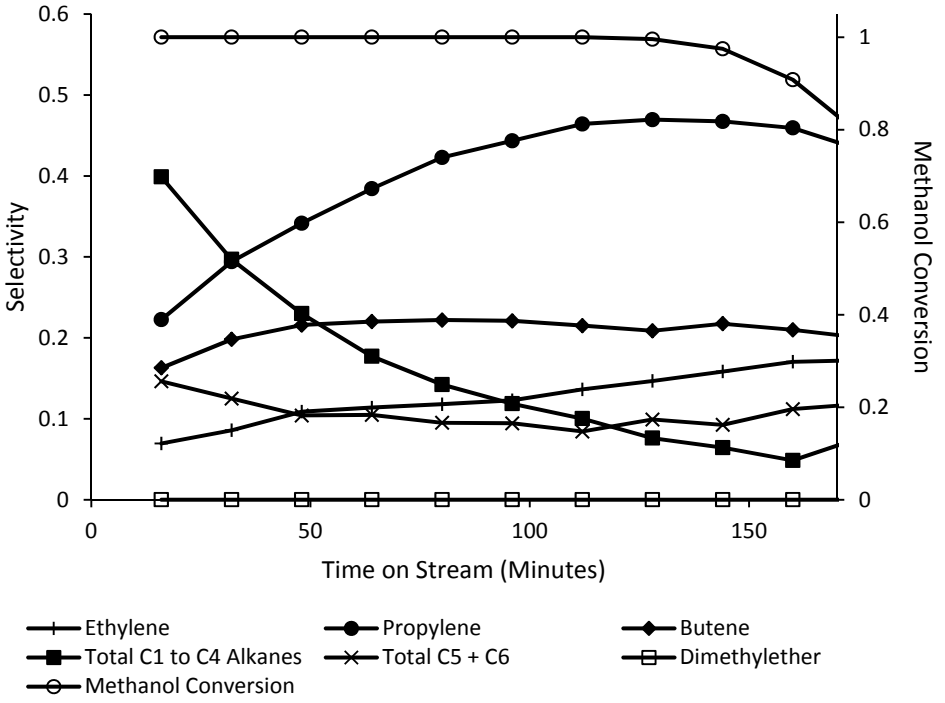

Figure S18. RTH (Si/Al=17)

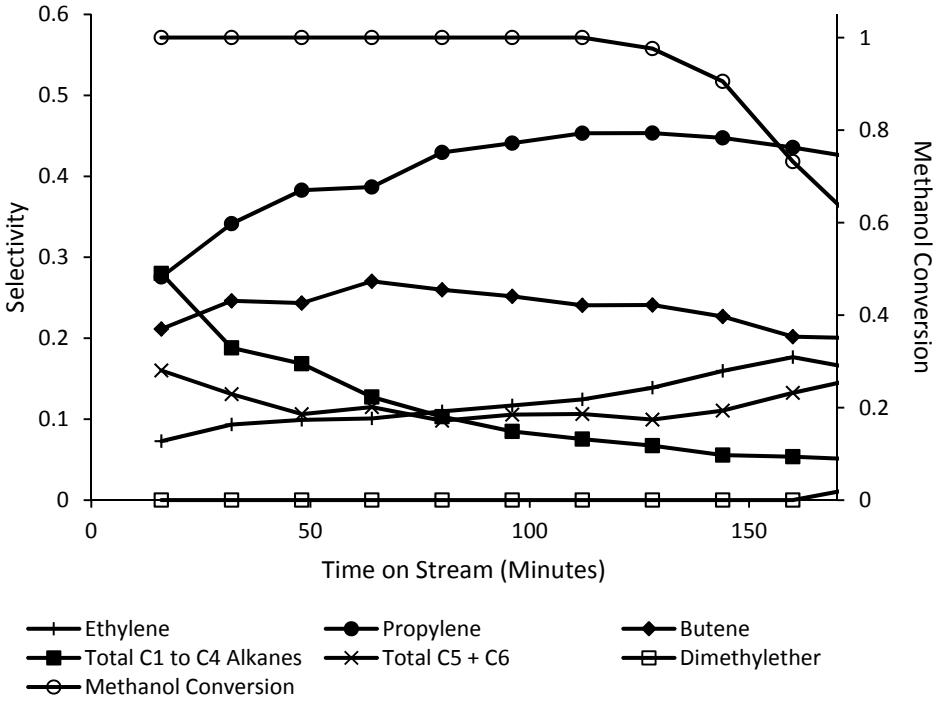

Figure S19. RTH (Si/Al=29)

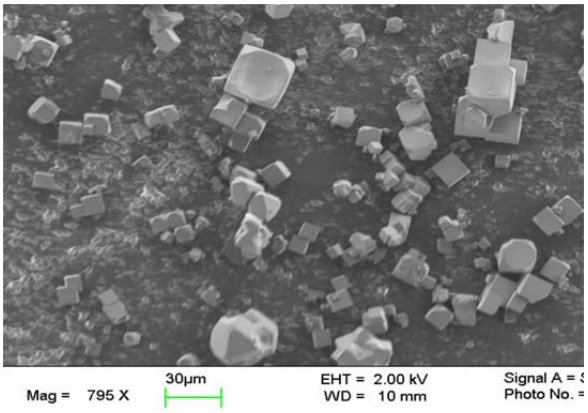

Figure S20. SEM images of pure-silica LTA

Crystal Structure Analysis of LTA

The crystal structure of three different LTA samples was solved using single crystal x-ray diffraction. The sample numbers and descriptions are below. Following this is the structural information for the three samples. Cif files for each sample are attached separately.

Table S2. Single crystal sample description

Sample Number	Description
p15133	As-made material, no TMA present in synthesis
p15134	As-made material, TMA present in synthesis
p15229	Calcined pure-silica LTA

Sample p15133

Table S3. Crystal data and structure refinement for p15133.

Identification code	p15133	
Empirical formula	F0.08 O2 Si	
Formula weight	61.69	
Temperature	100 K	
Wavelength	0.71073 Å	
Crystal system	Cubic	
Space group	Pm-3m	
Unit cell dimensions	a = 11.813(3) Å	a= 90°
	b = 11.813(3) Å	b= 90°
	c = 11.813(3) Å	g = 90°
Volume	1648.5(13) Å3	
Z	24	
Density (calculated)	1.491 Mg/m3	
Absorption coefficient	0.552 mm-1	
F(000)	738	
Crystal size	0.08 x 0.07 x 0.05 mm3	
Theta range for data collection	2.987 to 47.986°.	
Index ranges	-17<=h<=14, -19<=k<=24, -22<=l<=18	
Reflections collected	25900	
Independent reflections	1599 [R(int) = 0.0535]	
Completeness to theta = 25.000°	99.1 %	
Absorption correction	Semi-empirical from equivalents	
Max. and min. transmission	1.0000 and 0.8819	
Refinement method	Full-matrix least-squares on F2	
Data / restraints / parameters	1599 / 0 / 24	
Goodness-of-fit on F2	1.099	
Final R indices [I>2sigma(I)]	R1 = 0.0497, wR2 = 0.1145	
R indices (all data)	R1 = 0.0623, wR2 = 0.1188	
Extinction coefficient	n/a	
Largest diff. peak and hole	0.814 and -1.019 e.Å-3	

Table S4. Atomic coordinates (x 105) and equivalent isotropic displacement parameters (Å2x 104) for p15133. U(eq) is defined as one third of the trace of the orthogonalized Uij tensor.

	x	y	z	U(eq)
Si(1)	50000	87042(3)	31738(3)	21(1)
O(1)	50000	79664(8)	20336(8)	46(2)
O(2)	61343(6)	83950(9)	38657(6)	101(2)
O(3)	50000	100000	27463(12)	109(2)
F(1)	50000	100000	50000	108(8)

Table S5. Bond lengths [Å] and angles [°] for p15133.

Si(1)-O(1)	1.6043(6)
Si(1)-O(2)#1	1.6115(5)
Si(1)-O(2)	1.6115(6)
Si(1)-O(3)	1.6119(7)
O(1)-Si(1)#2	1.6043(6)
O(2)-Si(1)#3	1.6115(6)
O(3)-Si(1)#4	1.6119(7)
O(1)-Si(1)-O(2)	107.62(4)
O(1)-Si(1)-O(2)#1	107.62(4)
O(1)-Si(1)-O(3)	104.65(7)
O(2)-Si(1)-O(2)#1	112.50(8)
O(2)#1-Si(1)-O(3)	111.97(5)
O(2)-Si(1)-O(3)	111.97(5)
Si(1)-O(1)-Si(1)#2	155.81(9)
Si(1)#3-O(2)-Si(1)	142.39(7)
Si(1)#4-O(3)-Si(1)	143.49(10)

Symmetry transformations used to generate equivalent atoms:

#1 -x+1,y,z #2 x,-z+1,-y+1 #3 -z+1,y,-x+1

#4 -x+1,-y+2,z

Table S6. Anisotropic displacement parameters (Å2x 104) for p15133. The anisotropic displacement factor exponent takes the form: -2p2[h2 a*2U11 + ... + 2 h k a* b* U12]

	U11	U22	U33	U23	U13	U12
Si(1)	24(1)	17(1)	21(1)	-1(1)	0	0
O(1)	72(4)	33(2)	33(2)	-20(3)	0	0
O(2)	82(2)	139(4)	82(2)	-53(2)	-59(3)	53(2)
O(3)	237(7)	10(4)	79(5)	0	0	0
F(1)	101(10)	121(16)	101(10)	0	0	0

Table S7. Torsion angles [°] for p15133.

O(1)-Si(1)-O(2)-Si(1)#3	178.13(12)
O(1)-Si(1)-O(3)-Si(1)#4	180.000(1)
O(2)-Si(1)-O(1)-Si(1)#2	60.74(5)
O(2)#1-Si(1)-O(1)-Si(1)#2	-60.74(5)
O(2)#1-Si(1)-O(2)-Si(1)#3	-63.48(16)
O(2)#1-Si(1)-O(3)-Si(1)#4	63.71(4)
O(2)-Si(1)-O(3)-Si(1)#4	-63.71(4)
O(3)-Si(1)-O(1)-Si(1)#2	180.000(1)
O(3)-Si(1)-O(2)-Si(1)#3	63.66(15)

Symmetry transformations used to generate equivalent atoms:

#1 -x+1,y,z #2 x,-z+1,-y+1 #3 -z+1,y,-x+1

#4 -x+1,-y+2,z

Sample p15134

Table S8. Crystal data and structure refinement for p15134.

Identification code	p15134	
Empirical formula	C0.17 H0.50 F0.10 N0.04 O2 Si	
Formula weight	65.17	
Temperature	100 K	
Wavelength	0.71073 Å	
Crystal system	Cubic	
Space group	Pm-3m	
Unit cell dimensions	a = 11.824(5) Å	a= 90°
	b = 11.824(5) Å	b= 90°
	c = 11.824(5) Å	g = 90°
Volume	1653(2) Å3	
Z	24	
Density (calculated)	1.571 Mg/m3	
Absorption coefficient	0.557 mm-1	
F(000)	786	
Crystal size	0.09 x 0.09 x 0.08 mm3	
Theta range for data collection	2.436 to 51.444°.	
Index ranges	-22<=h<=24, -16<=k<=24, -25<=l<=25	
Reflections collected	52351	
Independent reflections	1863 [R(int) = 0.0646]	
Completeness to theta = 25.000°	99.4 %	
Absorption correction	Semi-empirical from equivalents	
Max. and min. transmission	1.0000 and 0.8917	
Refinement method	Full-matrix least-squares on F2	
Data / restraints / parameters	1863 / 0 / 31	
Goodness-of-fit on F2	1.123	
Final R indices [I>2sigma(I)]	R1 = 0.0296, wR2 = 0.0750	
R indices (all data)	R1 = 0.0427, wR2 = 0.0798	
Extinction coefficient	n/a	
Largest diff. peak and hole	0.581 and -0.512 e.Å-3	

Table S9. Atomic coordinates (x 105) and equivalent isotropic displacement parameters (Å2x 104) for p15134. U(eq) is defined as one third of the trace of the orthogonalized Uij tensor.

	x	y	z	U(eq)
Si(1)	50000	87120(2)	31734(2)	40(1)
O(1)	50000	79536(4)	20464(4)	64(1)
O(2)	61351(3)	83896(5)	38649(3)	87(1)
O(3)	50000	100000	27260(7)	90(1)
F(1)	50000	100000	50000	95(3)
N(1)	50000	50000	50000	94(4)
C(1)	50000	60270(50)	42760(50)	329(18)

Table S10. Bond lengths [Å] and angles [°] for p15134.

Si(1)-O(1)	1.6063(7)
Si(1)-O(2)#1	1.6172(7)
Si(1)-O(2)	1.6172(7)
Si(1)-O(3)	1.6122(8)
O(1)-Si(1)#2	1.6062(7)
O(2)-Si(1)#3	1.6172(7)
O(3)-Si(1)#4	1.6121(8)
N(1)-C(1)#5	1.486(6)
N(1)-C(1)#6	1.486(6)
N(1)-C(1)#7	1.486(6)
N(1)-C(1)#8	1.486(6)
N(1)-C(1)#9	1.486(6)
N(1)-C(1)#10	1.486(6)
N(1)-C(1)#11	1.486(6)
N(1)-C(1)#12	1.486(6)
N(1)-C(1)#3	1.486(6)
N(1)-C(1)#13	1.486(6)
N(1)-C(1)#14	1.486(6)
N(1)-C(1)#15	1.486(6)
C(1)-C(1)#2	0.508(13)
C(1)-C(1)#16	1.718(8)
C(1)-C(1)#6	1.711(13)
C(1)-C(1)#3	1.210(9)

C(1)-C(1)#5	1.529(6)
C(1)-C(1)#17	1.210(9)
C(1)-C(1)#7	1.718(8)
C(1)-C(1)#8	1.529(6)
C(1)-C(1)#18	1.529(6)
C(1)-C(1)#19	1.529(6)
C(1)-H(1B)	1.0853
C(1)-H(1A)	0.9333
C(1)-H(1C)	1.1512

O(1)-Si(1)-O(2)	106.73(2)
O(1)-Si(1)-O(2)#1	106.73(2)
O(1)-Si(1)-O(3)	104.78(4)
O(2)-Si(1)-O(2)#1	112.19(4)
O(3)-Si(1)-O(2)	112.87(2)
O(3)-Si(1)-O(2)#1	112.87(2)
Si(1)#2-O(1)-Si(1)	157.88(5)
Si(1)-O(2)-Si(1)#3	141.59(4)
Si(1)#4-O(3)-Si(1)	141.69(5)
C(1)#12-N(1)-C(1)#13	131.9(3)
C(1)#9-N(1)-C(1)#13	70.6(3)
C(1)#5-N(1)-C(1)#8	118.09(10)
C(1)#11-N(1)-C(1)#15	118.09(10)
C(1)#11-N(1)-C(1)#8	118.09(10)
C(1)#3-N(1)-C(1)#12	61.91(9)
C(1)#14-N(1)-C(1)#13	19.7(5)
C(1)#6-N(1)-C(1)#12	70.6(3)
C(1)#15-N(1)-C(1)#12	19.7(5)
C(1)#14-N(1)-C(1)#15	109.4(3)
C(1)#3-N(1)-C(1)#14	180.0
C(1)#7-N(1)-C(1)#15	160.3(5)
C(1)#3-N(1)-C(1)#10	61.91(10)
C(1)#10-N(1)-C(1)#12	61.91(10)
C(1)#14-N(1)-C(1)#10	118.09(10)
C(1)#7-N(1)-C(1)#12	180.0
C(1)#3-N(1)-C(1)#11	131.9(3)
C(1)#14-N(1)-C(1)#8	160.3(5)
C(1)#14-N(1)-C(1)#11	48.1(3)
C(1)#9-N(1)-C(1)#8	109.4(3)
C(1)#10-N(1)-C(1)#11	160.3(5)

C(1)#12-N(1)-C(1)#8	48.1(3)
C(1)#3-N(1)-C(1)#6	48.1(3)
C(1)#11-N(1)-C(1)#13	61.91(10)
C(1)#14-N(1)-C(1)#6	131.9(3)
C(1)#5-N(1)-C(1)#13	61.91(10)
C(1)#10-N(1)-C(1)#6	19.7(5)
C(1)#3-N(1)-C(1)#15	70.6(3)
C(1)#11-N(1)-C(1)#6	180.0(4)
C(1)#10-N(1)-C(1)#15	48.1(3)
C(1)#3-N(1)-C(1)#9	118.09(10)
C(1)#9-N(1)-C(1)#15	131.9(3)
C(1)#14-N(1)-C(1)#9	61.91(10)
C(1)#5-N(1)-C(1)#15	180.0
C(1)#10-N(1)-C(1)#9	180.0
C(1)#14-N(1)-C(1)#12	118.09(10)
C(1)#11-N(1)-C(1)#9	19.7(5)
C(1)#11-N(1)-C(1)#12	109.4(3)
C(1)#6-N(1)-C(1)#9	160.3(5)
C(1)#9-N(1)-C(1)#12	118.09(10)
C(1)#3-N(1)-C(1)#7	118.09(10)
C(1)#5-N(1)-C(1)#12	160.3(5)
C(1)#14-N(1)-C(1)#7	61.91(9)
C(1)#3-N(1)-C(1)#8	19.7(5)
C(1)#10-N(1)-C(1)#7	118.09(10)
C(1)#10-N(1)-C(1)#8	70.6(3)
C(1)#11-N(1)-C(1)#7	70.6(3)
C(1)#6-N(1)-C(1)#8	61.91(10)
C(1)#6-N(1)-C(1)#7	109.4(3)
C(1)#7-N(1)-C(1)#8	131.9(3)
C(1)#9-N(1)-C(1)#7	61.91(10)
C(1)#15-N(1)-C(1)#8	61.91(10)
C(1)#3-N(1)-C(1)#5	109.4(3)
C(1)#3-N(1)-C(1)#13	160.3(5)
C(1)#14-N(1)-C(1)#5	70.6(3)
C(1)#10-N(1)-C(1)#13	109.4(3)
C(1)#10-N(1)-C(1)#5	131.9(3)
C(1)#6-N(1)-C(1)#13	118.09(10)
C(1)#11-N(1)-C(1)#5	61.91(10)
C(1)#7-N(1)-C(1)#13	48.1(3)
C(1)#6-N(1)-C(1)#5	118.09(10)

C(1)#15-N(1)-C(1)#13	118.09(10)
C(1)#9-N(1)-C(1)#5	48.1(3)
C(1)#8-N(1)-C(1)#13	180.0
C(1)#7-N(1)-C(1)#5	19.7(5)
C(1)#6-N(1)-C(1)#15	61.91(10)
N(1)-C(1)-C(1)#8	59.04(5)
N(1)-C(1)-C(1)#6	54.8(3)
N(1)-C(1)-C(1)#19	59.04(5)
N(1)-C(1)-C(1)#18	59.04(5)
N(1)-C(1)-C(1)#16	54.68(12)
N(1)-C(1)-C(1)#5	59.04(5)
N(1)-C(1)-C(1)#7	54.68(13)
N(1)-C(1)-H(1B)	115.4
N(1)-C(1)-H(1A)	127.6
N(1)-C(1)-H(1C)	111.2
C(1)#17-C(1)-N(1)	65.97(16)
C(1)#2-C(1)-N(1)	80.2(3)
C(1)#3-C(1)-N(1)	65.97(16)
C(1)#2-C(1)-C(1)#17	119.998(5)
C(1)#19-C(1)-C(1)#16	16.7(4)
C(1)#19-C(1)-C(1)#7	80.4(2)
C(1)#2-C(1)-C(1)#7	59.999(3)
C(1)#7-C(1)-C(1)#16	90.0
C(1)#2-C(1)-C(1)#5	43.3(4)
C(1)#2-C(1)-C(1)#18	103.3(4)
C(1)#18-C(1)-C(1)#7	43.3(4)
C(1)#3-C(1)-C(1)#8	16.7(4)
C(1)#17-C(1)-C(1)#8	99.6(2)
C(1)#2-C(1)-C(1)#3	119.998(6)
C(1)#18-C(1)-C(1)#16	113.32(19)
C(1)#3-C(1)-C(1)#16	60.000(1)
C(1)#2-C(1)-C(1)#8	103.3(4)
C(1)#3-C(1)-C(1)#18	99.6(2)
C(1)#8-C(1)-C(1)#6	56.0(3)
C(1)#17-C(1)-C(1)#18	16.7(4)
C(1)#5-C(1)-C(1)#6	103.6(3)
C(1)#8-C(1)-C(1)#5	112.9(3)
C(1)#18-C(1)-C(1)#6	56.0(3)
C(1)#18-C(1)-C(1)#5	60.000(1)
C(1)#19-C(1)-C(1)#6	103.6(3)

C(1)#2-C(1)-C(1)#19	43.3(4)
C(1)#3-C(1)-C(1)#7	120.000(2)
C(1)#17-C(1)-C(1)#7	60.0
C(1)#3-C(1)-C(1)#19	76.7(4)
C(1)#8-C(1)-C(1)#7	113.32(19)
C(1)#17-C(1)-C(1)#19	124.19(7)
C(1)#5-C(1)-C(1)#7	16.7(4)
C(1)#3-C(1)-C(1)#5	124.19(7)
C(1)#6-C(1)-C(1)#7	90.000(2)
C(1)#8-C(1)-C(1)#19	60.000(1)
C(1)#17-C(1)-C(1)#16	120.000(1)
C(1)#18-C(1)-C(1)#19	112.9(3)
C(1)#8-C(1)-C(1)#16	43.3(4)
C(1)#17-C(1)-C(1)#5	76.7(4)
C(1)#5-C(1)-C(1)#16	80.4(2)
C(1)#3-C(1)-C(1)#17	90.0
C(1)#6-C(1)-C(1)#16	90.0
C(1)#3-C(1)-C(1)#6	45.001(1)
C(1)#17-C(1)-C(1)#6	45.001(1)
C(1)#8-C(1)-C(1)#18	105.2(3)
C(1)#5-C(1)-C(1)#19	68.1(6)
C(1)#2-C(1)-C(1)#6	134.999(2)
C(1)#2-C(1)-C(1)#16	59.999(3)
C(1)#8-C(1)-H(1B)	86.4
C(1)#17-C(1)-H(1B)	173.3
C(1)#16-C(1)-H(1B)	62.7
C(1)#18-C(1)-H(1B)	157.8
C(1)#7-C(1)-H(1B)	114.9
C(1)#5-C(1)-H(1B)	98.2
C(1)#3-C(1)-H(1B)	96.5
C(1)#19-C(1)-H(1B)	56.4
C(1)#6-C(1)-H(1B)	141.5
C(1)#2-C(1)-H(1B)	55.1
C(1)#7-C(1)-H(1A)	78.8
C(1)#19-C(1)-H(1A)	141.1
C(1)#5-C(1)-H(1A)	83.8
C(1)#8-C(1)-H(1A)	158.8
C(1)#2-C(1)-H(1A)	97.9
C(1)#18-C(1)-H(1A)	70.9
C(1)#16-C(1)-H(1A)	157.8

C(1)#3-C(1)-H(1A)	142.1
C(1)#17-C(1)-H(1A)	70.7
C(1)#6-C(1)-H(1A)	108.9
C(1)#2-C(1)-H(1C)	145.2
C(1)#16-C(1)-H(1C)	98.7
C(1)#6-C(1)-H(1C)	65.1
C(1)#3-C(1)-H(1C)	48.0
C(1)#19-C(1)-H(1C)	113.1
C(1)#17-C(1)-H(1C)	94.2
C(1)#7-C(1)-H(1C)	153.4
C(1)#8-C(1)-H(1C)	61.3
C(1)#18-C(1)-H(1C)	110.8
C(1)#5-C(1)-H(1C)	168.6
H(1B)-C(1)-H(1A)	104.6
H(1B)-C(1)-H(1C)	91.3
H(1A)-C(1)-H(1C)	99.8

Symmetry transformations used to generate equivalent atoms:

#1 -x+1,y,z #2 x,-z+1,-y+1 #3 -z+1,y,-x+1

#4 -x+1,-y+2,z #5 z,-x+1,-y+1 #6 x,y,-z+1

#7 -y+1,-x+1,z #8 y,-z+1,x #9 x,z,-y+1 #10 -x+1,-z+1,y

#11 -x+1,-y+1,z #12 y,x,-z+1 #13 -y+1,z,-x+1

#14 z,-y+1,x #15 -z+1,x,y #16 y,x,z #17 z,y,-x+1

#18 -y+1,-z+1,-x+1 #19 -z+1,-x+1,-y+1

Table S11. Anisotropic displacement parameters (Å2x 104) for p15134. The anisotropic displacement factor exponent takes the form: -2p2[h2 a*2U11 + ... + 2 h k a* b* U12]

	U11	U22	U33	U23	U13	U12
Si(1)	44(1)	35(1)	40(1)	-4(1)	0	0
O(1)	84(2)	54(1)	54(1)	-25(2)	0	0
O(2)	77(1)	108(2)	77(1)	-29(1)	-35(1)	29(1)
O(3)	161(3)	34(2)	75(2)	0	0	0
F(1)	101(4)	84(6)	101(4)	0	0	0
N(1)	94(4)	94(4)	94(4)	0	0	0
C(1)	680(50)	120(20)	190(20)	70(20)	0	0

Table S12. Hydrogen coordinates (x 104) and isotropic displacement parameters (Å2x 10 3) for p15134.

	x	y	z	U(eq)
H(1B)	5571	6003	3558	49
H(1A)	4375	6436	4021	49
H(1C)	5511	6747	4687	49

Table S13. Torsion angles [°] for p15134.

O(1)-Si(1)-O(2)-Si(1)#3	178.14(6)
O(1)-Si(1)-O(3)-Si(1)#4	180.000(1)
O(2)#1-Si(1)-O(1)-Si(1)#2	-60.07(3)
O(2)-Si(1)-O(1)-Si(1)#2	60.07(3)
O(2)#1-Si(1)-O(2)-Si(1)#3	-65.31(8)
O(2)#1-Si(1)-O(3)-Si(1)#4	64.26(2)
O(2)-Si(1)-O(3)-Si(1)#4	-64.26(2)
O(3)-Si(1)-O(1)-Si(1)#2	180.000(1)
O(3)-Si(1)-O(2)-Si(1)#3	63.56(8)
C(1)#11-N(1)-C(1)-C(1)#7	60.06(15)
C(1)#8-N(1)-C(1)-C(1)#19	71.33(4)
C(1)#12-N(1)-C(1)-C(1)#18	-128.0(5)
C(1)#9-N(1)-C(1)-C(1)#5	40.7(4)
C(1)#3-N(1)-C(1)-C(1)#16	69.20(7)
C(1)#15-N(1)-C(1)-C(1)#16	79.2(2)
C(1)#10-N(1)-C(1)-C(1)#19	139.3(4)
C(1)#14-N(1)-C(1)-C(1)#5	-10.0(3)
C(1)#5-N(1)-C(1)-C(1)#6	139.3(4)
C(1)#8-N(1)-C(1)-C(1)#5	152.8(7)
C(1)#3-N(1)-C(1)-C(1)#2	129.27(9)
C(1)#7-N(1)-C(1)-C(1)#19	-100.8(2)
C(1)#14-N(1)-C(1)-C(1)#2	-50.73(9)
C(1)#10-N(1)-C(1)-C(1)#6	0.000(2)
C(1)#10-N(1)-C(1)-C(1)#2	180.000(4)
C(1)#13-N(1)-C(1)-C(1)#6	112.1(3)
C(1)#11-N(1)-C(1)-C(1)#2	0.000(4)
C(1)#15-N(1)-C(1)-C(1)#7	-160.7(5)
C(1)#6-N(1)-C(1)-C(1)#2	180.000(4)
C(1)#6-N(1)-C(1)-C(1)#16	119.94(15)
C(1)#9-N(1)-C(1)-C(1)#2	0.000(4)
C(1)#5-N(1)-C(1)-C(1)#18	71.33(4)
C(1)#7-N(1)-C(1)-C(1)#2	-60.06(15)
C(1)#13-N(1)-C(1)-C(1)#18	44.2(6)
C(1)#5-N(1)-C(1)-C(1)#2	-40.7(4)
C(1)#11-N(1)-C(1)-C(1)#5	40.7(4)
C(1)#15-N(1)-C(1)-C(1)#2	139.3(4)
C(1)#15-N(1)-C(1)-C(1)#5	180.000(1)
C(1)#12-N(1)-C(1)-C(1)#2	119.94(15)

C(1)#3-N(1)-C(1)-C(1)#19	88.5(4)
C(1)#8-N(1)-C(1)-C(1)#2	112.1(3)
C(1)#6-N(1)-C(1)-C(1)#19	139.3(4)
C(1)#13-N(1)-C(1)-C(1)#2	-67.9(3)
C(1)#15-N(1)-C(1)-C(1)#19	98.5(7)
C(1)#14-N(1)-C(1)-C(1)#3	180.000(1)
C(1)#3-N(1)-C(1)-C(1)#6	-50.73(9)
C(1)#10-N(1)-C(1)-C(1)#3	50.73(9)
C(1)#9-N(1)-C(1)-C(1)#6	180.000(2)
C(1)#11-N(1)-C(1)-C(1)#3	-129.27(9)
C(1)#12-N(1)-C(1)-C(1)#6	-60.06(15)
C(1)#6-N(1)-C(1)-C(1)#3	50.73(9)
C(1)#14-N(1)-C(1)-C(1)#7	9.3(2)
C(1)#9-N(1)-C(1)-C(1)#3	-129.27(9)
C(1)#9-N(1)-C(1)-C(1)#7	60.06(15)
C(1)#7-N(1)-C(1)-C(1)#3	170.7(2)
C(1)#8-N(1)-C(1)-C(1)#7	172.14(16)
C(1)#5-N(1)-C(1)-C(1)#3	-170.0(3)
C(1)#10-N(1)-C(1)-C(1)#16	119.94(15)
C(1)#15-N(1)-C(1)-C(1)#3	10.0(3)
C(1)#7-N(1)-C(1)-C(1)#16	-120.1(3)
C(1)#12-N(1)-C(1)-C(1)#3	-9.3(2)
C(1)#8-N(1)-C(1)-C(1)#16	52.0(5)
C(1)#8-N(1)-C(1)-C(1)#3	-17.2(4)
C(1)#15-N(1)-C(1)-C(1)#18	-108.67(4)
C(1)#13-N(1)-C(1)-C(1)#3	162.8(4)
C(1)#8-N(1)-C(1)-C(1)#18	-135.8(6)
C(1)#3-N(1)-C(1)-C(1)#17	-101.47(17)
C(1)#3-N(1)-C(1)-C(1)#5	170.0(3)
C(1)#14-N(1)-C(1)-C(1)#17	78.53(17)
C(1)#10-N(1)-C(1)-C(1)#5	-139.3(4)
C(1)#10-N(1)-C(1)-C(1)#17	-50.73(9)
C(1)#6-N(1)-C(1)-C(1)#5	-139.3(4)
C(1)#11-N(1)-C(1)-C(1)#17	129.27(9)
C(1)#7-N(1)-C(1)-C(1)#5	-19.3(5)
C(1)#6-N(1)-C(1)-C(1)#17	-50.73(9)
C(1)#12-N(1)-C(1)-C(1)#5	160.7(5)
C(1)#9-N(1)-C(1)-C(1)#17	129.27(9)
C(1)#13-N(1)-C(1)-C(1)#5	-27.2(7)
C(1)#7-N(1)-C(1)-C(1)#17	69.20(7)

C(1)#14-N(1)-C(1)-C(1)#19	-91.5(4)
C(1)#5-N(1)-C(1)-C(1)#17	88.5(4)
C(1)#11-N(1)-C(1)-C(1)#19	-40.7(4)
C(1)#15-N(1)-C(1)-C(1)#17	-91.5(4)
C(1)#9-N(1)-C(1)-C(1)#19	-40.7(4)
C(1)#12-N(1)-C(1)-C(1)#17	-110.80(7)
C(1)#5-N(1)-C(1)-C(1)#19	-81.5(7)
C(1)#8-N(1)-C(1)-C(1)#17	-118.7(2)
C(1)#12-N(1)-C(1)-C(1)#19	79.2(2)
C(1)#13-N(1)-C(1)-C(1)#17	61.3(2)
C(1)#13-N(1)-C(1)-C(1)#19	-108.67(4)
C(1)#3-N(1)-C(1)-C(1)#8	17.2(4)
C(1)#14-N(1)-C(1)-C(1)#6	129.27(9)
C(1)#14-N(1)-C(1)-C(1)#8	-162.8(4)
C(1)#11-N(1)-C(1)-C(1)#6	180.000(2)
C(1)#10-N(1)-C(1)-C(1)#8	67.9(3)
C(1)#7-N(1)-C(1)-C(1)#6	119.94(15)
C(1)#11-N(1)-C(1)-C(1)#8	-112.1(3)
C(1)#15-N(1)-C(1)-C(1)#6	-40.7(4)
C(1)#6-N(1)-C(1)-C(1)#8	67.9(3)
C(1)#8-N(1)-C(1)-C(1)#6	-67.9(3)
C(1)#9-N(1)-C(1)-C(1)#8	-112.1(3)
C(1)#3-N(1)-C(1)-C(1)#7	-170.7(2)
C(1)#7-N(1)-C(1)-C(1)#8	-172.14(16)
C(1)#10-N(1)-C(1)-C(1)#7	-119.94(15)
C(1)#5-N(1)-C(1)-C(1)#8	-152.8(7)
C(1)#6-N(1)-C(1)-C(1)#7	-119.94(15)
C(1)#15-N(1)-C(1)-C(1)#8	27.2(7)
C(1)#5-N(1)-C(1)-C(1)#7	19.3(5)
C(1)#12-N(1)-C(1)-C(1)#8	7.86(16)
C(1)#12-N(1)-C(1)-C(1)#7	180.000(1)
C(1)#13-N(1)-C(1)-C(1)#8	180.000(1)
C(1)#13-N(1)-C(1)-C(1)#7	-7.86(16)
C(1)#3-N(1)-C(1)-C(1)#18	-118.7(2)
C(1)#14-N(1)-C(1)-C(1)#16	-110.80(7)
C(1)#14-N(1)-C(1)-C(1)#18	61.3(2)
C(1)#11-N(1)-C(1)-C(1)#16	-60.06(15)
C(1)#10-N(1)-C(1)-C(1)#18	-67.9(3)
C(1)#9-N(1)-C(1)-C(1)#16	-60.06(15)
C(1)#11-N(1)-C(1)-C(1)#18	112.1(3)

C(1)#5-N(1)-C(1)-C(1)#16	-100.8(2)
C(1)#6-N(1)-C(1)-C(1)#18	-67.9(3)
C(1)#12-N(1)-C(1)-C(1)#16	59.9(3)
C(1)#9-N(1)-C(1)-C(1)#18	112.1(3)
C(1)#13-N(1)-C(1)-C(1)#16	-128.0(5)
C(1)#7-N(1)-C(1)-C(1)#18	52.0(5)

Symmetry transformations used to generate equivalent atoms:

#1 -x+1,y,z #2 x,-z+1,-y+1 #3 -z+1,y,-x+1

#4 -x+1,-y+2,z #5 z,-x+1,-y+1 #6 x,y,-z+1

#7 -y+1,-x+1,z #8 y,-z+1,x #9 x,z,-y+1 #10 -x+1,-z+1,y

#11 -x+1,-y+1,z #12 y,x,-z+1 #13 -y+1,z,-x+1

#14 z,-y+1,x #15 -z+1,x,y #16 y,x,z #17 z,y,-x+1

#18 -y+1,-z+1,-x+1 #19 -z+1,-x+1,-y+1

Sample p15229

Table S14. Crystal data and structure refinement for p15229.

Identification code	p15229	
Empirical formula	O2 Si	
Formula weight	60.09	
Temperature	100 K	
Wavelength	0.71073 Å	
Crystal system	Cubic	
Space group	Pm-3m	
Unit cell dimensions	a = 11.857(4) Å	a= 90°
	b = 11.857(4) Å	b= 90°
	c = 11.857(4) Å	g = 90°
Volume	1667.0(17) Å3	
Z	24	
Density (calculated)	1.437 Mg/m3	
Absorption coefficient	0.540 mm-1	
F(000)	720	
Crystal size	0.10 x 0.10 x 0.10 mm3	
Theta range for data collection	2.429 to 60.181°.	
Index ranges	-14<=h<=27, -23<=k<=22, -27<=l<=23	
Reflections collected	48084	
Independent reflections	2479 [R(int) = 0.0861]	
Completeness to theta = 25.000°	99.7 %	
Absorption correction	Semi-empirical from equivalents	
Max. and min. transmission	1.0000 and 0.8962	
Refinement method	Full-matrix least-squares on F2	
Data / restraints / parameters	2479 / 0 / 21	
Goodness-of-fit on F2	1.152	
Final R indices [I>2sigma(I)]	R1 = 0.0449, wR2 = 0.0833	
R indices (all data)	R1 = 0.0850, wR2 = 0.0933	
Extinction coefficient	n/a	
Largest diff. peak and hole	1.530 and -0.750 e.Å-3	

Table S15. Atomic coordinates (x 105) and equivalent isotropic displacement parameters (Å2x 104) for p15229. U(eq) is defined as one third of the trace of the orthogonalized Uij tensor.

	x	y	z	U(eq)
Si(1)	50000	86918(2)	31655(2)	38(1)
O(1)	50000	79613(4)	20387(4)	68(1)
O(2)	61064(4)	84036(6)	38936(4)	136(1)
O(3)	50000	100000	28299(7)	139(2)

Table S16. Bond lengths [Å] and angles [°] for p15229.

Si(1)-O(1)	1.5922(6)
Si(1)-O(2)#1	1.6072(6)
Si(1)-O(2)	1.6072(6)
Si(1)-O(3)	1.6014(6)
O(1)-Si(1)#2	1.5922(6)
O(2)-Si(1)#3	1.6072(6)
O(3)-Si(1)#4	1.6014(6)
O(1)-Si(1)-O(2)	109.57(3)
O(1)-Si(1)-O(2)#1	109.57(3)
O(1)-Si(1)-O(3)	108.57(4)
O(2)#1-Si(1)-O(2)	109.43(5)
O(3)-Si(1)-O(2)#1	109.84(3)
O(3)-Si(1)-O(2)	109.84(3)
Si(1)-O(1)-Si(1)#2	155.91(5)
Si(1)#3-O(2)-Si(1)	146.27(4)
Si(1)-O(3)-Si(1)#4	151.22(6)

Symmetry transformations used to generate equivalent atoms:

#1 -x+1,y,z #2 x,-z+1,-y+1 #3 -z+1,y,-x+1

#4 -x+1,-y+2,z

Table S17. Anisotropic displacement parameters (Å2x 104) for p15229. The anisotropic displacement factor exponent takes the form: -2p2[h2 a*2U11 + ... + 2 h k a* b* U12]

	U11	U22	U33	U23	U13	U12
Si(1)	48(1)	31(1)	34(1)	-7(1)	0	0
O(1)	98(3)	52(1)	52(1)	-28(2)	0	0
O(2)	116(1)	176(3)	116(1)	-56(2)	-76(2)	56(2)
O(3)	289(5)	24(2)	103(3)	0	0	0

Table S18. Hydrogen coordinates (x 104) and isotropic displacement parameters (Å2x 10 3)

for p15229.

	x	y	z	U(eq)

Table S19. Torsion angles [°] for p15229.

O(1)-Si(1)-O(2)-Si(1)#3	176.06(8)
O(1)-Si(1)-O(3)-Si(1)#4	180.000(1)
O(2)-Si(1)-O(1)-Si(1)#2	60.03(3)
O(2)#1-Si(1)-O(1)-Si(1)#2	-60.04(3)
O(2)#1-Si(1)-O(2)-Si(1)#3	-63.78(11)
O(2)#1-Si(1)-O(3)-Si(1)#4	60.20(3)
O(2)-Si(1)-O(3)-Si(1)#4	-60.20(3)
O(3)-Si(1)-O(1)-Si(1)#2	180.000(1)
O(3)-Si(1)-O(2)-Si(1)#3	56.88(10)

Symmetry transformations used to generate equivalent atoms:

#1 -x+1,y,z #2 x,-z+1,-y+1 #3 -z+1,y,-x+1

#4 -x+1,-y+2,z

References

(1) Pophale, R.; Daeyaert, F.; Deem, M. W. Computational Prediction of Chemically Synthesizable Organic Structure Directing Agents for Zeolites. *J. Mater. Chem. A* **2013**, *1*, 6750–6760.

(2) Corma, A.; Rey, F.; Rius, J.; Sabater, M. J.; Valencia, S. Supramolecular Self-Assembled Molecules as Organic Directing Agent for Synthesis of Zeolites. *Nature* **2004**, *431*, 287–290.